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Doran R. Pace, Patent Attorney

REQUEST FOR CERTIFICATE OF
CORRECTION UNDER 37 CFR 1.322
Docket No. UF.420XC1

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants : Swadeshmukul Santra, Paul H. Holloway, Robert A. Mericle, Heesun
Yang, Glenn A. Walter
Issued : March 6, 2012
Patent No. : 8,128,908
Serial No. : 10/590,590
Conf. No. : 8520
For : Nanoparticles and Their Use for Multifunctional Bioimaging

Mail Stop Certificate of Corrections Branch
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

REQUEST FOR CERTIFICATE OF CORRECTION
UNDER 37 CFR 1.322 (OFFICE MISTAKE)

Sir:

A Certificate of Correction for the above-identified patent has been prepared and is attached
hereto.

In the left-hand column below is the column and line number where errors occurred in the
patent. In the right-hand column is the page and line number in the application where the correct
information appears.

Patent Reads:

Column 31, Line 22:

“(ph 7)”

Application Reads:

Page 46, line 15:


--(ph~7)--

Column 32, Lines 41-42: $\text{“Mn}^{2+} 4\text{T}_1\text{-}^6\text{A}_1\text{”}$ Page 48, line 9: $\text{--Mn}^{2+} 4\text{T}_1\text{-}^6\text{A}_1\text{--.}$

A true and correct copy of pages 46 and 48 of the specification as filed which support Applicants' assertion of the errors on the part of the Patent Office accompanies this Certificate of Correction.

Approval of the Certificate of Correction is respectfully requested.

Respectfully submitted,



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DRP/ps

Attachments: Copy of pages 46 and 48 of the specification
Certificate of Correction

UNITED STATES PATENT AND TRADEMARK OFFICE
CERTIFICATE OF CORRECTION

Page 1 of 1

PATENT NO. : 8,128,908
APPLICATION NO. : 10/590,590
ISSUE DATE : March 6, 2012
INVENTOR(S) : Swadeshmukul Santra, Paul H. Holioway, Robert A. Mericle, Heesun Yang, Glenn A. Walter

It is certified that error appears in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

Column 31.

Line 22, "(ph 7)" should read --(ph~7)--.

Column 32.

Lines 41-42, " $\text{Mn}^{2+} 4\text{T}_1-^6\text{A}_1$ " should read -- $\text{Mn}^{2+} 4\text{T}_1-^6\text{A}_1$ --.

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Silica coating and Subsequent Surface-Functionalization of Quantum Dots. After addition of the Zn micellar solution, 2.5 ml of tetraethyl orthosilicate (TEOS) is injected into CdS:Mn/ZnS micellar solution and mixed for 15 min at room temperature. The hydrolysis of TEOS and condensation reaction is initiated by adding NH₄OH in the form of micellar solution, which is prepared by mixing 1.5 ml of NH₄OH (30 wt %) with AOT (3.69 g)/heptane (82.5 ml) stock solution. After polymerization for 24 hr at room temperature, 1.25 ml of TEOS and 0.25 ml of 3-(Aminopropyl)triethoxysilane (APTS) are injected into above solution and mixed for 10 min. And then, another NH₄OH micellar solution (prepared by mixing 0.9 ml of NH₄OH with AOT (2.21 g)/heptane (50 ml) stock solution) and 3-(Trihydroxysilyl)propyl methylphosphonate (THPMP) (prepared by mixing 0.75 ml of THPMP and 3.6 ml of water with AOT (4.42 g)/heptane (50 ml) stock solution) are injected subsequently and reacted for 24 hours. And silica-overcoated-, surface-functionalized quantum dots are precipitated by addition of a small amount of methanol. After thorough washing with methanol, these quantum dots are solubilized stably in a sodium phosphate buffer solution (pH \sim 7). ✓

Characterization. A JEOL JSM 6400 electron microscope operated at 15 kV was used for EDS analysis. A Perkin-Elmer PHI 5100 x-ray photoelectron spectrometer and Mg K_α x-ray (1253.6 eV) were used for XPS/ESCA. Survey scans were collected from 1100 to 0 eV with a step of 0.5 eV, a time/step of 30 ms, and a pass energy of 89.45 eV. Multiplex scans were collected with a step of 0.1 eV, a time/step of 50 ms, and pass energy of 35.75 eV. A JEOL 2010F transmission electron microscope operated at 200 kV was used for imaging and direct determination of the nanocrystal size. The XRD patterns were obtained with a Philips MRD X'Pert system for information on structure and crystal size. XRD pattern was collected in the step scan mode, typically with a scan range of 15° – 80°, a step of 0.01°, and a glazing angle of incident x-ray of 1°.

Quantum yields of *n*-dodecanethiol- and ZnS-passivated CdS:Mn nanocrystals dispersed in chloroform were determined by comparing the integrated emission to those from Coumarin 6, Coumarin 30, and perylene at the excitation wavelength of 385 nm. Coumarin 6, Coumarin 30, and perylene were dissolved in ethanol, acetonitrile, and cyclohexane, respectively. The optical densities of the sample and reference solutions were 0.065 ± 0.003 at the excitation wavelength.

resolution images of a nanocrystal with the zinc blende structure and the $(1\bar{1}0)$ and (110) planes are identified.

Example 2—Photoluminescence (PL) and Quantum Yield

5 PL emission spectra, obtained using 325 nm HeCd laser excitation, of *n*-dodecanethiol-passivated and ZnS-passivated CdS:Mn nanocrystals are compared in Figure 3A and Figure 3B, respectively. Note that PL measurements were carried out in the solid-state sample, *i.e.*, ~200 nm thick CdS:Mn/ZnS nanocrystal layers on the glass substrate. The $\text{Mn}^{2+} {}^6\text{T}_1 \rightarrow {}^6\text{A}_1$ transition at ~600 nm is observed from both nanocrystalline ✓
10 samples. In addition to the Mn emission at ~600 nm, emission from a surface-related defect (shallow trap) is observed at ~450 nm from organically passivated nanocrystals. This defect emission originates from the localized surface states in the band gap, which presumably are formed by the lack of bonding to surface S ions. In contrast, no defect-related emission is observed from inorganically passivated nanocrystals, indicating the
15 successful complete passivation of CdS:Mn core surface by the ZnS shell layer.

The quantum yields of CdS:Mn nanocrystals with either a *n*-dodecanethiol or ZnS capping layer were measured in chloroform solutions and found to be 3.7 and 18 %, respectively. Note that for quantum yield measurement, CdS:Mn/ZnS core/shell nanocrystals were capped with *n*-dodecanethiol in order to be soluble in chloroform. The
20 value of 3.7 % for organically capped CdS:Mn is reasonable since it is close to the quantum yields reported from organically passivated ZnS:Mn nanocrystals (1-4 %) (Kubo, T., T. Isobe, M. Senna, *J. Lumin.* 2002, 99:39; Bol, A., A. Meijerink, *J. Phys. Chem. B* 2001, 105, 10197). The enhanced quantum yield of *n*-dodecanethiol-capped CdS:Mn/ZnS nanocrystals is a direct result of more effective surface passivation, by
25 which nonradiative recombination paths are reduced significantly. A visual comparison of organically and inorganically passivated nanocrystals is shown in Figure 4. Both samples are standing on a handheld-UV lamp providing 366 nm multiband irradiation, and it is obvious that the ZnS-passivated nanocrystals are much brighter.

Example 3—Effects of UV irradiation and Photooxidation

30 The change of PL emission intensity from organically and inorganically passivated CdS:Mn during 400 nm UV irradiation was monitored at room temperature using a